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Review

Occurrence and dissipation mechanism of organic pollutants during the composting of sewage sludge: A critical review

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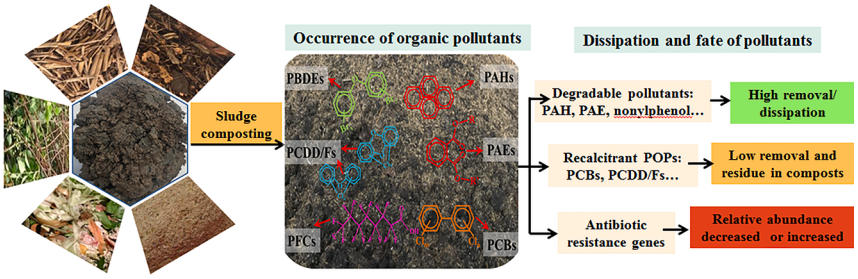
HIGHLIGHTS

- PAHs and PAEs showed higher dissipation percentages than PCDD/Fs and PCBs during sludge composting.
- Organic pollutants like PAHs and PAEs were removed mainly through degradation or mineralization.
- Recalcitrant PCDD/Fs and PCBs can be sequestrated in organic fraction of sludge compost.
- ARG abundance in some cases increased after sludge composting.
- Innovative and efficient composting is necessary to remove recalcitrant POPs and ARG.

GRAPHICAL ABSTRACT



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ABSTRACT

Sewage sludge contains various classes of organic pollutants, limiting its land application. Sludge composting can effectively remove some organic pollutants. This review summarizes recent researches on concentration changes and dissipation of different organic pollutants including persistent organic pollutants during sludge composting, and discusses their dissipation pathways and the current understanding on dissipation mechanism. Some organic pollutants like PAHs and phthalates were removed mainly through biodegradation or mineralization, and their dissipation percentages were higher than those of PCDD/Fs and PCBs. Nevertheless, some recalcitrant organic pollutants could be sequestrated in organic fractions of sludge mixtures, and their levels and ARG abundance even increased after sludge composting in some studies, posing potential risks for land application. This review demonstrated that microbial community and their corresponding degradation for organic pollutants were influenced by different pollutants, bulking agents, composting methods and processes. Further research perspectives on removing organic pollutants during sludge composting were highlighted.

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1. Introduction

Sewage sludge is generated with huge volume from wastewater treatment plant (Pan et al., 2018). At present, the main representative disposal ways of sewage sludge include landfill, land application, drying-incineration, composting, and recycling as building materials (Verlicchi and Zambello, 2015; Guo et al., 2020). Land application is a promising method of disposing sewage sludge (Verlicchi and Zambello, 2015). Nevertheless, sewage sludge contains pathogens, inorganic (e.g., heavy metals) and organic toxic pollutants that limit its land application (Fijalkowski et al., 2017; Dubey et al., 2021).

Recently, many studies have focused on organic pollutants in sewage sludge (Clarke and Smith, 2011; Meng et al., 2016; Fijalkowski et al., 2017; Guo et al., 2020; Dubey et al., 2021). The organic pollutants of great concern include polyaromatic hydrocarbons (PAHs), polychlorinated biphenyl (PCBs), polychorodibenzo-p-dioxins and polychorodibenzofurans (PCDD/Fs), polybrominated diphenyl ethers (PBDEs), phthalates (PAEs), and perfluorinated compounds (PFCs) (Harrison et al., 2006; Clarke and Smith, 2011; Meng et al., 2016; Fijalkowski et al., 2017; Sun et al., 2019). Pharmaceuticals and personal care products (PPCPs, especially antibiotics) in sewage sludge have also attracted the increasing concern (Verlicchi and Zambello, 2015; Ezzariai et al., 2018). The levels of some organic pollutants such as PCDD/Fs in sludge from Spain and German (Eljarrat et al., 1999; Zennegg et al., 2013) and PAHs in sludge from China (Meng et al., 2016) generally showed a decline trend, while those of PBDEs increased annually by 16% from Swedish (Olofsson et al., 2012). The maximum sum concentrations of 16 priority PAHs (Σ16PAHs) and di(2-ethylhexyl) phthalate (DEHP) in sewage sludge was up to 280 mg/kg dry weight (unless otherwise mentioned) (Sun et al., 2019) and 3514 mg/kg (Meng et al., 2014), respectively, exceeding the standard limits (6 mg/kg for Σ16PAHs; 100 mg/kg for DEHP) suggested by the European commission for land application. Land application of sludge can elevate the levels of organic pollutants in soils, runoff and groundwater (Verlicchi and Zambello, 2015; Kim et al., 2017). Therefore, it is necessary to lower the levels of hazardous pollutants in sewage sludge.

Composting is a cost-effective technology to decline or eliminate pathogens and organic pollutants in sewage sludge (Cai et al., 2007b, 2012; Ezzariai et al., 2018; Guo et al., 2020). Numerous studies were conducted to optimize composting methods and processes of sewage sludge, and to investigate their effect mechanism on pollutant removal. To 2020, more than 1500 papers have been published on sludge compost (searching both “compost” and “sludge” in Title of publications within Web of Science of Thomson). Since 2000, many studies have focused on the occurrence and fate of organic pollutants during sludge composting, and found that composting can promote the dissipation of organic pollutants (Cai et al., 2007b, 2012; Siebielska and Sidełko, 2015; Ezzariai et al., 2018; Guo et al., 2020). Recently, some studies have investigated the dissipation of emergent organic pollutants and their influencing factors (especially microbial community) during sludge composting (Yang et al., 2017; Alves et al., 2019). With the increasing number of studies on sludge composting and organic pollutants of concern, it is helpful to summarize the dissipation of organic pollutants and its mechanism as well as effect factors on their removals.

The objectives of this work are to (1) summarize the occurrence and removals of organic pollutants in sludge composts; (2) discuss the possible dissipation mechanisms of organic pollutants during sludge composting; (3) analyze the effect factors on removals of organic pollutants, especially on degradation by microbial community. Finally, research gaps and further perspectives on removing organic pollutants are addressed.

2. Occurrence and removal of organic pollutants in sludge composts

Recently, many efforts have been made on optimizing materials and methods for composting of sewage sludge to produce soil amendment and promote the removals or elimination of pathogens and contaminants (Fig. 1). Most of composting (82%) was performed at laboratory scale and only 18% was conducted at pilot/plant scale (e.g., windrowing) with various bulking agents (Fig. 1 and Table 1) (Ezzariai et al., 2018; Guo et al., 2020). Conventional thermophilic composting (maximum temperature 55–70 °C) was widely conducted (Cai et al., 2007b, 2012; Aemig et al., 2019), while hyperthermophilic composting (maximum temperatures up to 90 °C) was conducted in a few cases (Liao et al., 2018; Cui et al., 2020a). The variation in composting materials, methods and duration might lead to the differences in occurrence, distribution, and removals of organic contaminants during sludge composting, so it is difficult to compare the removal efficiency of organic pollutants among different studies. Therefore, this work mainly focuses on loading and residual concentrations of organic contaminants, dissipation/removal efficiency and mechanism, as well as effect factors (especially microbial community) in sludge composting.

2.1. Polycyclic aromatic hydrocarbons (PAHs)

Among various organic pollutants, PAHs are among the most frequently investigated pollutants in sludge composting. The reported PAH concentrations and removal efficiency during sludge composting are summarized in Table 1. The initial concentrations of Σ16PAHs in the mixture of sludge with bulking agents were generally below 30 mg/kg, even less than 5.0 mg/kg. After composting, the Σ16PAHs were generally below 10 mg/kg, even less than 1.0 mg/kg. Several studies have reported that the removal percentages of Σ16PAHs during sludge composting could reach more than 90%, while in some cases, those were between 80% and 90%, even less than 50% (Table 1). These results suggested that composting could partly remove PAHs in sludge. Differently, Lazzari et al. (2000) observed the increase of Σ16PAHs in maturity phase of sludge composting due to the mass reduction (around 40–60%) and low degradation/volatilization rates of some high molecular weight PAHs.

The dissipation of PAHs during sludge composting was influenced by sludge pretreatment methods (Lu et al., 2019), bulking agents (Rorat et al., 2017), composting methods (Poluszyńska et al., 2017), bioavailability of PAHs (Lu et al., 2019), and microbial degradation (Guo et al., 2020). Lu et al. (2019) reported that bioleaching conditioning enhanced the removal of PAHs after 39 d of sludge composting, with the dissipation percentages of bioleached sludge (76.4%) > Fe[III]/CaO-conditioned sludge (60.4%) > raw sludge (58.7%). Regarding various bulking agents, the removal percentages of Σ16PAHs in sludge vermicomposting for five weeks decreased in the order of market wastes (85.8%) > green waste (61.6%) > organic fraction of municipal solid waste (57.9%) (Rorat et al., 2017). The variation in PAH removal efficiency among different pretreated sludge or bulking agents may be attributed to the differences in bioavailability of PAHs in sludge mixtures and in the composition/abundance of potential PAH-degraders (Oleszczuk, 2009; Lu et al., 2019; Guo et al., 2020). Bioleached sludge might enhance bioavailability of individual PAHs during composting (Lu et al., 2019). Good linear correlations were observed between the PAH removal percentages and their bioavailability during sludge composting (Oleszczuk, 2009; Lu et al., 2019). Nevertheless, partial adsorption of PAHs to non-accessible sites (e.g., lignocellulose, holocellulose) temporarily limited their mobility and allowed them to escape microbial attack (Amir et al., 2005a; Aemig et al., 2019), resulting in low removal of PAHs.

The dissipation of PAHs during sludge composting was also affected by the temperature (Amir et al., 2005a). The conventional aerobic composting generally experienced four phases, namely mesophilic, thermophilic, cooling and maturation (Amir et al., 2005a; Guo et al., 2020). The decrease of PAH concentrations during sludge composting possibly resulted from the combination of degradation and volatilization (Hua et al., 2008). In some studies the major PAH dissipation was observed in mesophilic and thermophilic phases (Lazzari et al., 2000;

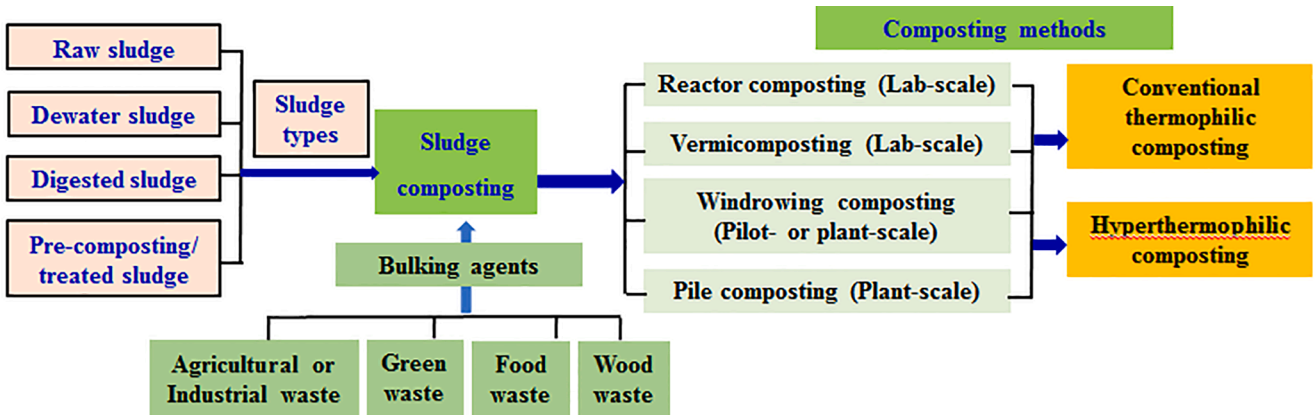

Fig. 1. Various types of sludge composting.

Table 1
PAH concentrations and removal after sewage sludge composting.

PAHs	Bulking agent (%)	Composting time (d)	Initial concentration (mg/kg)	Final concentration (mg/kg)	Removal efficiency (%)	References
∑PAHs	Green forest waste [a]	50	5.58–9.83	1.81–2.88	62–75	Guo et al., 2020
∑PAHs	Mushroom residue [b]	39	13.5–15.9	–	58.7–76.4	Lu et al., 2019
Benzo(*a*)pyrene	Sawdust/mushroom residue [b]	60	5	–	51.1	Liu et al., 2019
∑PAHs	Food and industry wastes [a]	365	82 ± 25 ng/g	83 + 33 ng/g	57 ± 31	Ozaki et al., 2017
Benzo(*a*)pyrene	Food and industry wastes [a]		4.3 ± 2.1 ng/g	2.4 ± 2.8 ng/g	76 ± 29	Ozaki et al., 2017
∑PAHs	Sawdust [a]	30	10.1–25.3	1.34–5.21	84.7	Poluszyńska et al., 2017
Benzo(*a*)pyrene	Sawdust [a]	30	<0.04–1.30	~0.52		Poluszyńska et al., 2017
∑PAHs	Green waste (55%) [b]	35	2.94	1.16	61.6	Rorat et al., 2017
Benzo(*a*)pyrene	Green waste (55%) [b]	35	0.37	0.028	92.5	Rorat et al., 2017
∑PAHs	Waste from markets (30%) [b]	35	3.96	0.56	85.8	Rorat et al., 2017
Benzo(*a*)pyrene	Waste from markets (30%) [b]	35	1.65	0.056	96.6	Rorat et al., 2017
∑PAHs	Bark and peat [a]	124	8.0	0.6	92.5	Kapanen et al., 2013
∑PAHs	Sawdust [a]	56	18.1	5.62–8.20	54.6–75.9	Cai et al., 2012
Benzo(*a*)pyrene	Sawdust [a]	56	1.02	~0.28	56.7–72.1	Cai et al., 2012
∑PAHs	Green waste [a]	28	0.93 ± 0.02	1.53 ± 0.03	+	Patureau et al., 2012
∑PAHs	Green waste (2:1) [a]	90	0.48		67.8–100[f]	Hafidi et al., 2008
Benzo(*a*)pyrene	Green waste (2:1) [a]	90	0.024	<0.001	99.5	Hafidi et al., 2008
∑PAHs	Rapeseed marc (10%) [a]	50	75.2	16.2	79.0	Hua et al., 2008
Benzo(*a*)pyrene	Rapeseed marc (10%) [a]	50	1.75	0.63	63.9	Hua et al., 2008
∑PAHs	- [b]	76	3.67–11.2		29.3–46.8	Oleszczuk, 2008
Benzo(*a*)pyrene	- [b]	76	0.24–1.27	0.11–0.38		Oleszczuk, 2008
∑PAHs	Straw [a]	56	28.6	1.8–10.2	64–94	Cai et al., 2007a, 2007b
Benzo(*a*)pyrene	Straw [a]	56	0.62	0.06–1.1	~90	Cai et al., 2007a, 2007b
∑PAHs	Fly ash (20%) [b]	353	6.96 ± 0.57	1.15 ± 0.10	83.4	Oleszczuk, 2006
Benzo(*a*)pyrene	Fly ash (20%) [b]	353	0.25 ± 0.01	0.16 ± 0.01	46.3	Oleszczuk, 2006
∑PAHs	Sawdust [b]	353	8.25 ± 0.68	0.98 ± 0.09	88.1	Oleszczuk, 2006
Benzo(*a*)pyrene	Sawdust [b]	353	0.36 ± 0.04	0.17 ± 0.02	52.6	Oleszczuk, 2006
∑PAHs	Straw (11%) [a]	180	0.21	0.02	88	Amir et al., 2005a, 2005b
∑PAHs	Green and ligneous wastes [a]	90	0.7	0.93–1.0	+	Lazzari et al., 2000

[a] The composting was in pilot-scale.
[b] The composting was in lab-scale.

Kopec et al., 2017), while those were mainly found in the stabilization phase of sludge composting by Amir et al. (2005a) and Hafidi et al. (2008). These differences in PAH removals might be related to the variation in PAH bioavailability and decomposition of organic matter (Lazzari et al., 1999). The highest decomposition of organic matter in sludge was accompanied with the greatest PAH removal (Kopec et al., 2017).

Regarding individual PAH compounds after sludge composting, their residual concentrations were generally below 10 mg/kg (Cai et al., 2007b; Hua et al., 2008), even less than 0.1 mg/kg (Amir et al., 2005a; Ozaki et al., 2017). The residual Σ16PAHs was mainly dominated by 3-ring PAHs (Hua et al., 2008) or 3-, 4- ring PAHs (Ozaki et al., 2017), while the high molecular weight PAHs (⩾ 4 rings) contributed low percentage to Σ16PAHs (Cai et al., 2007b; Ozaki et al., 2017). In general, phenanthrene, fluoranthene, and pyrene displayed relatively higher concentrations compared with other PAH compounds (Amir et al., 2005a; Ozaki et al., 2017). The removal of individual PAH compounds depend on their initial concentrations, aromatic rings, and composting methods. For example, Poluszyńska et al. (2017) observed that both sludge vermicomposting and composting with sawdust could degrade 2–6 ring PAHs with removal percentages of 100% for 2-ring, 88–94% for 3-ring, 64.4–74.4% for 4-ring, 80.6–83.2% for 5-ring, and 86.9–100% for 6-ring PAHs, which was far higher than those of fluoranthene, benzo [b]fluoranthene and benzo[a]pyrene (<50%) in the composting (66 d) of anaerobic digested sludge (Aemig et al., 2019). Even elevated levels for high molecular weight PAHs (⩾ 4 rings) and phenanthrene were observed in sludge composting (Hafidi et al., 2008; Ozaki et al., 2017). These increasing concentrations might be attributed to their tight sorption/sequestration or formation of bound-residue which made them inaccessible for microbial attack (Amir et al., 2005a; Hafidi et al., 2008).

Benzo[a]pyrene, the most toxic PAH compound with five aromatic rings, can influence the sludge composting process and the land

application of compost produced (Liu et al., 2019). The concentrations of benzo[a]pyrene in sludge composts were generally < 1.0 mg/kg (Table 1). Most studies found that composting could effectively remove benzo[a]pyrene by 46.3–99.5% (Table 1). However, our previous study demonstrated that, after 56 d of sludge composting, the concentrations of benzo[a]pyrene increased in the treatments of manually turned compost and inoculated-manual turned compost (Cai et al., 2007b). This might be related to the variation in pile temperature, benzo[a]pyrene bioavailability, and microbial community among different composting treatments (Cai et al., 2007b; Liu et al., 2019). More efficient composting method is necessary for the removal of benzo[a]pyrene in view of its notorious carcinogenic toxicity.

Some studies concluded that the dissipation of PAHs during sludge composting was mainly attributed to the microbial degradation (Amir et al., 2005a; Lu et al., 2019; Liu et al., 2019; Guo et al., 2020). During the composting process, specific conditions could enhance the microbial diversity and activity. Some PAH-degrading genera (e.g., Pseudomonas, Bacillus, Rhodococcus, and Sphingomonas) were observed in the sludge compost (Lu et al., 2019; Guo et al., 2020). For example, within the first 12 d of sludge composting (spiked with benzo(a)pyrene or not), the typical thermophilic bacteria such as Bacilli and Clostridia were the predominant class (Liu et al., 2019). In sludge composting with green forest wastes, the main genera including Bacillus, Pseudomonas and Methylotenera capable of degrading cellulose and PAH co-metabolizing contributed remarkable to PAH degradation (Guo et al., 2020). Obviously, PAHs in sludge compost can influence the structure and diversity of microbial communities (Liu et al., 2019) that correspondingly influence PAH degradation.

2.2. Polychlorinated dibenzo-p-dioxins and polychorodibenzofurans (PCDD/Fs)

PCDD/Fs are listed as persistent organic pollutants (POPs) in Stockholm Convention (Martinez et al., 2007). The study on PCDD/Fs in sewage sludge and its compost is limited due to the high cost for analysis of PCDD/Fs. Significant decline (more than one order of magnitude) for the total I-TEQ (international toxicity equivalent) levels of PCDD/Fs was observed in sewage sludge from Spain and Germany since the late 1970 s to 2000 s (Eljarrat et al., 1999; Martinez et al., 2007). In China, the total levels of PCDD/Fs in sewage sludge from 13 provinces were between 104 and 1661 pg/g and 2.51–75.2 pg I-TEQ/g (Lu et al., 2012), in few cases higher than Chinese limit (100 pg/g) proposed for land application.

Several studies have investigated the occurrence and distribution of PCDD/Fs in sludge composts (Table 2). In sludge composting mixed with straw and sawdust from a composting plant in Spain, the levels of PCDD/Fs varied from 1.38 to 117 ng/g (7.4–300 pg TEQ-WHO/g) (Muñoz et al., 2018). The concentrations of PCDD/Fs in composted sludge (~44.4 ng I-TEQ/kg) might be lower than those in thermal dried sludge (~91.7 ng I-TEQ/kg) due to the solid dilution with the addition of other materials (Martinez et al., 2007). In most cases, PCDD/F levels in composts were below the suggested limit (100 ng TEQ/kg) by European Commission (Martinez et al., 2007; Beníšek et al., 2015; Muñoz et al., 2018).

Regarding PCDD/F congeners, octachlorodibenzo-p-dioxin (OCDD) and 1,2,3,4,6,7,8-heptachlorodibenzo-p-dioxin (HpCDD) were the predominant congeners in sludge compost (Beníšek et al., 2015; Muñoz et al., 2018). HpCDD, OCDD and octachorodibenzofuran (OCDF) contributed 90–99% to the total concentrations of PCDD/Fs (Muñoz et al., 2018). The level of HpCDD was about 10-fold greater than that for OCDF. The highest OCDD concentrations (30–90 ng/g) were observed in three samples collected from the composting plant (Muñoz et al., 2018). OCDD levels in the final compost were greater than the initial concentrations in the mixture of sludge with straw and sawdust (Muñoz et al., 2018). This might be attributed to two reasons: (1) a concentration of recalcitrant PCDD/Fs with the mass reduction during sludge composting; (2) the formation of OCDD (80–1500 pg/g) at laboratory composting of sludge spiked with pentachlorophenol and non-forced aeration (Muñoz et al., 2018). It should be pointed out that no study reported the removal or degradation percentage for PCDD/Fs during sludge composting, which might be attributed to the elevated levels of dominant congeners (e.g., OCDD). Effective composting methods are necessary to be developed for reducing the residue of PCDD/Fs during sludge composting.

2.3. Polychlorinated biphenyls (PCBs)

PCBs are listed as POPs in Stockholm Convention and has been banned/restricted for decades. PCB concentrations in sewage sludge displayed a decreasing trend with apparent half-life between 9 and 12 years (Zennegg et al., 2013). The total concentrations of seven PCBs (Σ7PCBs, 5.6 to 11.0 mg/kg) in sewage sludge from Egypt (Barakat et al., 2017) far higher than those in other countries such as Switzerland, Spain, and Australia (Zennegg et al., 2013).

Several studies demonstrated the occurrence and distribution of PCBs in sludge composts where the ΣPCBs was generally less than 1 mg/kg (Table 2). Recalcitrant PCBs is difficult to be degraded during the composting, resulting in the lower removal (Gibson et al., 2007;

Table 2
Concentrations of PCDD/Fs and PCBs in sludge composts.

Compounds	Bulking agent	Composting time	Initial concentration	Final concentration		References
	(%)	(d)	(μg/kg)	(μg/kg)	(pg I-TEQ/g)	
PCDD/Fs	Sawdust and straw [a]	40		1.38–39.7	7.4–300	Muñoz et al., 2018
OCDD	Sawdust and straw [a]	40		1.0–34.9		Muñoz et al., 2018
PCDD/Fs	Various compost plants			0.04–0.47	2.9–15.1 [c]	Beníšek et al., 2015
PCDD/Fs	Treated by biomethanation	21–30	0.049	0.84–1.52	4.7–5.95	Muñoz et al., 2014
PCDD/Fs	Green waste and sifting refuse [a]	28	6.0–11.1 ng/kg	12.2–15 ng/kg		Patureau et al., 2012
PCDD/Fs	Pine wood chips [b]	75	15,600	16,000		Ramírez et al., 2008
PCDD/Fs	Pilot and field scale	26–124	3.8 pg I-TEQ/g		Nd-3.6	Kapanen et al., 2013
PCBs	Pilot and field scale	26–124	40–1400	200–700		Kapanen et al., 2013
PCBs	Pine wood chips [b]	75	34	15		Ramírez et al., 2008
PCBs	Various compost plants			0.98–44	14–130[c]	Beníšek et al., 2015
PCBs	Municipal waste [b]	182		<1.0	6.9–15.7	Siebielska and Sidełko, 2015
PCBs	Treated by biomethanation	21–30	0.50	3.77–14.1	1.01–1.55[c]	Muñoz et al., 2014
PCBs	Pine wood chips [b]	75	34	29		Ramírez et al., 2008
PCBs	Straws [a]	143	137,134	124,116		Gibson et al., 2007
PCBs	Green and ligneous [a]	90	128 ± 6	78 ± 7		Lazzari et al., 2000

[a] The composting was in pilot-scale.
[b] The composting was in lab-scale.
[c] The unit was pg TEQ-WHO/g.

Siebielska and Sidełko, 2015) and even increased PCB concentrations (Lazzari et al., 1999). For example, Gibson et al. (2007) reported that the ΣPCBs decreased from 136 to 120 μg/kg after 143 d of sludge composting with 11.7% of removal. Similar removal of PCBs (about 7–16%) was observed in the composting (182 d) of sludge with municipal waste (Siebielska and Sidełko, 2015). Nevertheless, contradictory research results were observed in the literature. The ΣPCBs did not decrease but instead increased by 37% during the whole composting process (Lazzari et al., 1999, 2000). The low removal or even increase of PCBs might be partly related to the strong sorption of PCBs onto organic matter during sludge composting and mass reduction (Lazzari et al., 2000). In view of the toxicity of recalcitrant PCB, PCB biodegradation, keystone degrading genera and removal pathway during sludge composting is needed to be investigated.

2.4. Phthalic acid esters (PAEs)

PAEs are widely applied as plasticizers and in non-polymeric products, and frequently detected in sewage sludge (Cai et al., 2007a; Meng et al., 2014). The concentrations of PAEs in sludge generally varied from several to hundreds (mg/kg), and the highest level was 58300 mg/kg (Harrison et al. 2006). Obviously, the levels of PAEs were far greater than those of PAHs, PCBs, and PCDD/Fs in sewage sludge. DEHP and di-*n*-butyl phthalate are the predominant PAE compounds in sewage sludge (Cai et al., 2007a; Fijalkowski et al., 2017). In certain cases DEHP levels exceeded the limit (100 mg/kg) suggested by European Commission for land application of sludge (Cai et al., 2007a; Meng et al., 2014), posing potential risk to ecosystem.

In sludge compost, six priority PAEs, in most cases only DEHP, were investigated (Cai et al., 2007c; Gibson et al., 2007; Poulsen and Bester, 2010). The concentrations of predominant DEHP in final compost were generally < 100 mg/kg (Table 3), while those of di-*n*-butyl phthalate were < 5.0 mg/kg (Cai et al., 2007c; Fu et al., 2013). The levels of other priority PAEs such as butyl benzyl phthalate and di-*n*-octyl phthalate were generally < 2.0 mg/kg (Cai et al., 2007b, 2012; Fu et al., 2013).

The biodegradability of PAEs is higher than that of PCBs and PCDD/Fs. Sludge composting could significantly decrease the levels of PAEs, with PAE removal percentages from 32.2% to 90.6% (Table 3). For DEHP, its removal percentages could reach 90% even up to 91–99% in sludge composting, while in some cases were < 50% (Table 3). DEHP degradation in sludge composting fitted a first-order kinetic model with a rate of 0.55–3.54 × 10^{-2} d^{-1}, which was higher than its biodegradation during mesophilic anaerobic digestion (about 0.23 × 10^{-2} d^{-1}) (Amir et al., 2005b). The composting methods and bulking agents had influences on the removal of PAEs from sewage sludge, as confirmed by more effective removal from sludge composting with pig manure than that with rice straw, especially with intermittent aeration (Fu et al., 2013). Similarly, intermittent aeration led to higher removal percentages of PAEs than the manual-turned composting (Cai et al., 2007c). This might be attributed to an increase in efficiency of oxygen supplementation and high abundance and diversity of indigenous microbial communities (Fu et al., 2013), which accelerated the dissipation of PAEs from sludge compost.

The degradation pathway of PAEs includes β-oxidation of alkyl side-chains, de-esterification, and transesterification during sludge composting (Amir et al., 2005b; Fu et al., 2013). For example, the degradation of DEHP involved in de-esterification (hydrolysis or dealkylation of side-chain) to form mono-ethyl-hexyl phthalate which could be further biotransformed into phthalic acid (Amir et al., 2005b; Fu et al., 2013). Di-*n*-butyl phthalate was degraded through β-oxidation of alkyl side-chains (the main catabolic pathway) to form diethyl phthalate (Amir et al., 2005b). After 30 d of sludge composting, di-*n*-butyl phthalate disappeared with the occurrence of diethyl phthalate and dimethyl phthalate; then diethyl phthalate vanished but the concentrations of dimethyl phthalate kept almost stable in sludge composts (Amir et al., 2005b). Furthermore, the levels of dimethyl phthalate increased all the time (Fu et al., 2013). Unfortunately, very limited information is available on the verification of PAE biodegradation by microbial community during sludge composting, even though some bacterial and fungi strains isolated from sludge were capable of degrading PAEs efficiently (Zhao et al., 2018).

2.5. Nonylphenol and nonylphenol ethoxylates

Nonylphenol with endocrine disrupting effect is mainly used as the raw material to produce nonylphenol ethoxylates which is the most widely used non-ionic surfactants. Nonylphenol and nonylphenol ethoxylates present in different sludge, and their sum levels in 78–100% samples of sludge and composts were higher than the limit (50 mg/kg) recommended by European Union for land application (González et al., 2010)

Some studies have investigated the levels and removal of nonylphenol and nonylphenol ethoxylates during sludge composting (Gibson et al., 2007; Zheng et al., 2018). During sludge composting, the degradation of nonylphenol ethoxylates could form nonylphenol (Pakou et al., 2009; González et al., 2010). González et al. (2010) observed

Table 3
PAE concentrations and removal percentages after sludge composting.

PAEs	Bulking agents (%)	Composting time (d)	Initial levels (mg/kg)	Final levels (mg/kg)	Removal efficiency (%)	References
∑PAEs	Pig manure [b]	60	25.5	5.60–6.45	74.7–78.0	Fu et al., 2013
DEHP	Pig manure [b]	60	22.5	2.84–5.20	76.9–87.4	Fu et al., 2013
∑PAEs	Rice straw [b]	60	25.5	13.9–17.3	32.2–45.5	Fu et al., 2013
DEHP	Rice straw [b]	60	22.5	12.7–16.1	28.4–43.6	Fu et al., 2013
∑PAEs	Rice straw [a]	56	–	9.80–18.0	58.3–90.6	Cai et al., 2007a, 2007b
DEHP	Rice straw [a]	56	–	8.10–16.0	~57.8	Cai et al., 2007a, 2007b
DEHP	Green waste [a]	28	6.4 ± 1	8.1 ± 0.2	+	Patureau et al., 2012
DEHP	Yard and park waste [a]	24	31.0	26.0	84.0	Poulsen and Bester, 2010
DEHP	Manure [b]	62	55.1	1.64	97.0	Pakou et al., 2009
DEHP	Sawdust [a]	18	200–300	<100.0	85.0–88.0	Cheng et al., 2008
DEHP	- [a]	143	36.4	13.2	63.7	Gibson et al., 2007
DEHP	Straw [a]	180	6.26	0.56	91.0	Amir et al., 2005b
DEHP	Grass [a]	135	28.7	1.72	94.0	Amir et al., 2005b
DEHP	- [a]	–	62.0–99.0	38.0–59.0	38.7–40.4	Bagó et al., 2005
DEHP	Tree bark and peat [a]	85	77.0	50.8	34.0	Marttinen et al., 2004
DEHP	Garden park waste [b]	25	38.0–160	<5.00	96.0–99.0	Moeller and Reeh, 2003
DEHP	Straw and green waste [b]	25	3.00–6.00	<1.00	91.0	Moeller and Reeh, 2003
DEHP	Shredded shrubbery [a]	370	–	–	86.0	Hartlieb et al., 2001

[a] The composting was in pilot-scale.
[b] The composting was in lab-scale.

higher concentrations of nonylphenol (mean: 179 mg/kg) and the sum of (nonylphenol + nonylphenol ethoxylates) in sludge composts than in sludge. On the other hand, a considerable removal (64–95%) of nonylphenol and nonylphenol ethoxylates were recorded during some sludge composting (Pakou et al., 2009; Zheng et al., 2018). The removal percentages of nonylphenol and nonylphenol ethoxylates depended on their initial concentrations in compost mixture (Pakou et al., 2009), bulking agents and mesophilic phase (Zheng et al., 2018). The removal percentages of nonylphenol (71.6%) in sludge composted with 20% mushroom residue was far higher than that (22.5%) with 20% sawdust (Zheng et al., 2018).

Composting conditions (e.g., ventilation and pH value) and their microbial community can influence the degradation and removal of nonylphenol. Increasing ventilating time or high-volume aeration promoted the degradation of nonylphenol during sludge composting (Pakou et al., 2009; Zheng et al., 2018). The removal percentage of nonylphenol was elevated from 19.7% to 41.6% with prolonged ventilation from 5 to 15 min during composting (Zheng et al., 2018). Anaerobic degradation efficiency of nonylphenol in sludge was promoted at alkaline condition (pH 10, removal: 47.0–69.4%), being almost 2-fold higher than that of the control (without pH adjustment, removal: 24.6%) within 8 d (Duan et al., 2018). A study demonstrated that certain specific microbial genera or families (e.g., Bacillus, *Sphingomonas*, *Pseudomonas*, *Alicycliphilus*) were capable of degrading nonylphenol, with significant correlation between nonylphenol dissipation percentage and the abundance of Bacillus (Zheng et al., 2018). High-volume aeration with high abundance of Bacillus (accounting for 60.9%) enhanced the activity and prolonged the survival of Bacillus, resulting in higher removal of nonylphenol compared to low-volume aeration (Zheng et al., 2018).

2.6. Pharmaceuticals and personal care products (PPCPs) and antibiotic resistance genes (ARGs)

PPCPs have received increasing concerns as emerging pollutants, especially antibiotics (Verlicchi and Zambello, 2015; Ezzariai et al., 2018). Various PPCP compounds (e.g., 152 pharmaceuticals and 17 personal care products) were observed in both untreated and treated sludge (e.g., digested, dewatered, conditioned, and dried sludge), with the concentrations ranging from 10^{-1} to 10^{5} ng/g (Verlicchi and Zambello, 2015). For example, the highest concentrations in primary sludge were observed for the fragrance galaxolide (187 μg/g) and tonalide (183 μg/g), while those in treated sludge were found for triclocarban (441 μg/g), tonalide (427 μg/g), galaxolide (177 μg/g), and triclosan (133 μg/g) (Verlicchi and Zambello, 2015). The concentrations of antibiotics in sewage sludge varied between 1 μg/kg and 100 mg/kg (Ezzariai et al., 2018). Land application of sewage sludge implies the elevating potential risk on the increasing PPCP levels in the environment and might lead to ARG dissemination (Verlicchi and Zambello, 2015; Ezzariai et al., 2018). Therefore, it is very urgent to propose strategies for managing and reducing the levels of PPCPs and limiting the dissemination of ARGs (Ezzariai et al., 2018).

Composting is a promising technique for partially removing some antibiotics and other PPCPs (Verlicchi and Zambello, 2015; Ezzariai et al., 2018). The most investigated classes of PPCPs in composted sludge were analgesics/anti-inflammatories and psychiatric drugs, followed by antibiotics, hormones and lipid regulators (Verlicchi and Zambello, 2015). After sludge composting, the levels of PPCPs reduced, but these compounds still remained in the final compost (Martín et al., 2012; Verlicchi and Zambello, 2015; Zheng et al., 2020). For example, the highest concentrations (1795 μg/kg) of ibuprofen in sludge composts were significantly lower than those in primary (14732 μg/kg), secondary (8450 μg/kg) and digested sludge (4223 μg/kg) (Martín et al., 2012). Significant dissipation was observed for PPCPs after composting of digested sludge (e.g., 98.2% and 74.8% removal for naproxen and 17β-estradiol, respectively) (Martín et al., 2012). High removal percentages (88.1–99.9%) were recorded for spiked pharmaceuticals (e.g., triclosan, diclofenac, ibuprofen, and carbamazepine) in the composting (92 d) of anaerobic sludge with waste wood (Butkovskyi et al., 2016). However, relative low removal (35–60%) of triclosan or even increase level was found in sludge composting (Ozaki et al., 2017; Zheng et al., 2020). Verlicchi and Zambello (2015) even listed triclosan, galaxolide tonalide (up to 10^{3} ng/g), and ibuprofen (close to 10^{3} ng/g) as the most recalcitrant substances in composted sludge. Triclosan could be partly transformed into recalcitrant methyltriclosan under mesophilic (12 ± 5%) and thermophilic (24 ± 10%) phases, which accumulated in composts (Butkovskyi et al., 2016). C/N ratio and ventilation influenced PPCP removal (Iranzo et al., 2018; Zheng et al., 2020). Among five C/N ratio treatments (17, 20, 24, 29, and 37), the removal of telmisartan was observed only in high C/N ratio (37) treatment, whereas fluoxetine, venlafaxine, and citalopram were removed in C/N ratio (20) treatment (Iranzo et al., 2018). High ventilation enhanced triclosan dissipation (59.5%) than low ventilation (48.1%) due to enough oxygen and high microorganism activity (Zheng et al., 2020). Notably, more efficient composting or removal methods are needed to be developed for degrading triclosan (methyltriclosan) and other recalcitrant PPCPs in sludge.

Concerning antibiotics, four groups of antibiotics (i.e., tetracyclines, fluoroquinolones, macrolides and sulfonamides) were focused due to their frequency occurrence in sewage sludge and manure (Ezzariai et al., 2018). Till now, about 10 studies were performed on the fate of antibiotic and ARGs during sludge composting, whereas more than 50 studies were conducted on the fate of antibiotic and ARGs during manure composting. Some studies found that antibiotics could be efficiently removed after composting of sludge (Lillenberg et al., 2010; Yang et al., 2016; Ezzariai et al., 2018). The concentrations (~22 μg/kg) of antibiotics (i.e., ciprofloxacin, norfloxacin, and sulfadimethoxine) in compost of sludge with tree bark were lower by one order of magnitude than those (~426 μg/kg) in sewage sludge (Lillenberg et al., 2010). Composting of sludge or manure could efficiently remove tetracyclines, sulfonamides, penicillin, and macrolides with dissipation percentages of 70–99% (Yang et al., 2016; Ezzariai et al., 2018). The dissipation of some antibiotics depended on composting method/phases and specific-compounds (Ezzariai et al., 2018; Khadra et al., 2019), with clarithromycin dissipation mainly at the first stage (0–15 d, 43%), lincomycin and tetracyclines at maturation stage (35–120 d, 36% and 75% respectively), while ampicillin and trimethoprim dissipated during all composting process (Khadra et al., 2019). High dissipation percentages (~99%) were found for enrofloxacin and norfloxacin in some studies, while others found that norfloxacin, ciprofloxacin and ofloxacin were recalcitrant to be biodegraded/transformed (Ezzariai et al., 2018). The elimination half-life of fluoroquinolone (217–2500 d) was longer than that of tetracyclines (1–350 d), macrolides (2–105 d) and sulfonamides (2–133 d) during sludge/manure composting (Ezzariai et al., 2018).

ARG accompanied with antibiotics in sludge and its compost (Ezzariai et al., 2018; Liao et al., 2018; Ren et al., 2018; Cui et al., 2020a, 2020b). The most focused ARGs are the resistance to tetracyclines (e.g., tetA, tetB, tetC, tetG), sulfonamides (e.g., sul1, sul2, sul3), macrolides (e.g., ermB, ermF, ermT, ermX), and fluoroquinolones (e.g., gyrA, parC, qnrA, qnrC, qnrS) (Ezzariai et al., 2018). Inconsistent results of decrease or increase in ARG were reported during sludge or manure composting (Ezzariai et al., 2018). Some studies observed the significant augment in the abundance of ARGs during sludge composting (Ezzariai et al., 2018; Liao et al., 2018; Ren et al., 2018). For example, the absolute abundance of ARGs increased about 75–740 folds after composting (66 d) of sludge with furfural slag compared with the initial sample (Ren et al., 2018). Total ARG copies were enriched by 2.04 and 1.95 folds in sludge composting with spent mushrooms and nitrification inhibitor (Zhang et al., 2016a), respectively. Optimistically, the relative abundance of ARGs decreased, even with high elimination percentages of ARGs in some sludge composting (Ezzariai et al., 2018; Liao et al., 2018; Ren et al., 2018; Cui et al., 2018, 2020a). The abundance of qnrA and qnrS genes was significantly reduced by 85.6–100% and 92.3–95.3%, respectively,

during vermicomposting of municipal excess sludge (Cui et al., 2018). It should be noted that ARG removal and fate were influenced by additives (e.g., biochar and zero-valent iron), pretreatments, and operational parameters (e.g., C/N ratio and composting temperature) (Liao et al., 2018; Cui et al., 2020b). Higher initial C/N ratio (30:1) could extend the thermophilic period and promote the removal of some target ARGs compared to the relative low C/N ratio (20:1 and 25:1) (Wei et al., 2020). Sludge hyperthermophilic composting removed ARGs and mobile genetic elements more effectively than conventional composting (89% vs 49%) (Liao et al., 2018). Semi-permeable membrane-covered thermophilic composting of sludge was also more efficient at decreasing the total abundances of ARGs and mobile genetic elements (decreasing by 42.1% and 38.1%) than conventional thermophilic composting (increasing by 4.5- and 1.6-folds) (Cui et al., 2020a). Reduction of mobile genetic elements played a crucial role in ARG removal during sludge hyperthermophilic composting (Liao et al., 2018).

Moreover, ARG patterns during sludge composting were positively correlated with bacterial community structure and abundance (Su et al., 2015; Liao et al., 2018). Sludge vermicomposting or hyperthermophilic composting could alter the bacterial community composition and decrease bacterial abundance and activity, which promoted the removal of ARGs and mobile genetic elements (Cui et al., 2018, 2020b; Liao et al., 2018). Nevertheless, distinct shifts were observed in structures and compositions of bacterial community during composting, even with large proportion of *Actinobacteria*, which might partially explain the ARG increase during sludge composting (Su et al., 2015). In view of the residual antibiotics and the increasing ARG abundance in some composting, more optimal and efficient composting process is necessary to avoid the dissemination of ARGs after land application of sludge compost.

2.7. Other organic pollutants

Several studies investigated the occurrence and removal of different kinds of organic micropollutants (Poulsen and Bester, 2010; Aemig et al., 2019). For example, Poulsen and Bester (2010) investigated 12 common organic micropollutants (including bactericides, flame retardants, fragrances, vulcanizers, and plasticizers) and found significant removal for 7 of the 12 micropollutants during sludge composting. The highest removal percentages of 13 organic pollutants (e.g., DEHP, tri-n-butyl phosphate, triclosan) was up to 99% (mean: 66%) during sludge composting (Sadef et al., 2014). Nevertheless, most studies investigated only one class of organic pollutants.

Linear alkylbenzene sulfonates (LAS, the anionic surfactants) have received concern earlier than many classes of organic contaminants in sludge. LAS concentrations in sewage sludge were relatively high (90–30900 mg/kg) (Pakou et al., 2009). Several studies investigated the LAS distribution and degradation during sludge composting (Moeller and Reeh, 2003; Pakou et al., 2009; Lashermes et al., 2012). Sludge composting (60 d) decreased significantly LAS concentrations, from 49.7 to 1490 mg/kg reduced to 24.8–297 mg/kg with removal percentages from 77.3% to 90.8% (Pakou et al., 2009). Faster (within 25 d) and higher removal (99%) of LAS was recorded in sludge composting conducted by Moeller and Reeh (2003). However, limited information is available on microbial community in composting sludge and their correlations with LAS degradation.

Bisphenols and tetrabromobisphenol A (and their alternatives) are widely used in daily consumer products and industrial productions, yet they exhibit endocrine disrupting effects. Bisphenol A and tetrabromobisphenol A were detected frequently with high concentrations in sewage sludge (Pilnáček et al., 2019; Dubey et al., 2021). Sludge composting could remove partly and lead to transformation of bisphenol A and tetrabromobisphenol A (Zhang et al., 2017; Pilnáček et al., 2019; Yang et al., 2017). For example, composting (14 d) of sewage sludge with sawdust could remove 50% of bisphenol A within the first 2.5–7.5 d (Zhang et al., 2017). Thermophilic aerobic degradation was the main removal pathway of bisphenol A (Zhang et al., 2017). Mesophilic treatment (i.e., biodrying) resulted in higher removal of bisphenol A than composting (88.3% vs 60.1%), which might be related to significant higher biomass of *Actinobacteria* (involving in degradation of bisphenol A) in the former than in the latter (Pilnáček et al., 2019). For tetrabromobisphenol A, sludge composting with spent mushroom compost could enhance the dissipation of tetrabromobisphenol A with significant higher removal percentages by 41.4–97.0% than in the composting of sludge alone (Yang et al., 2017). This might be related to spent mushroom compost containing enzymes, bacteria, and fungi as well as the presence of Geobacter and Mycobacterium involved in degradation of tetrabromobisphenol A (Yang et al., 2017). Tetrabromobisphenol A was almost completely transformed to bisphenol A via reductive debromination within 55 d by some dehalogenating bacteria (e.g., *Dehalococcoides*, and *Desulfovibrio*) in bench-scale anaerobic sludge reactors (Lefevre et al., 2016). Nevertheless, there is no report about the debromination of tetrabromobisphenol A during sludge composting.

PBDEs and PFCs are ubiquitous in sewage sludge and listed in Stockholm Convention (Yan et al., 2012; Kim et al., 2017; Dubey et al., 2021). Kim et al. (2017) reported that the highest concentration of PBDEs in sludge was up to 23500 ng/g until 2004–2007, and then decreased to 6600 ng/g, while Zennegg et al. (2013) found no clear temporal trend for the levels of PBDEs and long-chain perfluorinated acids in sewage sludge. The total concentrations of 14 perfluorinated acids in sewage sludge from Shanghai varied from 126 to 809 ng/g (mean: 452 ng/g) (Yan et al., 2012). The concentration of perfluorooctane sulfonate in sludge was even up to 3.12 mg/kg (Clarke and Smith, 2011). Unfortunately, because PBDEs and PFCs are recalcitrant, very few microbial genera were capable of promoting biodegradation or biotransformation PBDEs and PFCs (Yu et al., 2018a; Huang et al., 2019). No study to date has investigated the biodegradation (or biotransformation) and dissipation for PBDEs and PFCs during sludge composting. Abril et al. (2018) found that the levels of perfluorooctanoic acid and perfluorobutanoic acid in the compost of dehydrated sludge were higher than those in sludge. Therefore, the distribution and fate need to be investigated for PBDEs and PFCs during sludge composting.

3. Dissipation pathways and mechanism of organic pollutants during sludge composting

Significant advances were achieved on dissipation pathway and mechanism of organic pollutants during sludge composting (Ezzariai et al., 2018; Aemig et al., 2019). Organic pollutants could dissipate through biodegradation or mineralization as well as abiotic loss such as volatilization and lixiviation (or leaching) (Fig. 2). At the same time, some organic pollutants could be sequestrated through sorption or stabilization with organic fractions (Aemig et al., 2019). The dissipation or sequestration of organic pollutants during sludge composting were influenced by their physiochemical properties, composting methods (e.g., bulking agents, C/N ratio) and processes (including mesophilic and thermophilic temperature as well as duration), and bioavailability (Oleszczuk, 2009; Aemig et al., 2019; Lu et al., 2019). For some organic pollutants, especially the one with relatively high Henry law constant (e.g., $>10^{-4}$ atm-m^3/mol for PAH compounds), volatilization could contribute to their dissipation at high temperature during sludge composting (Gibson et al., 2007), while some organic contaminants (e.g., DEHP) were strongly adsorbed to solid matter of compost and thus its volatilization was negligible (Poulsen and Bester, 2010; Sadef et al., 2014). Additionally, the dissipation of halogenated organic compounds especially PFCs is generally lower than those of aromatic hydrocarbons (e.g., PAHs) during sludge composting (Tables 1–3). Short side-chain organic pollutants generally displayed higher removal than the long ones, or the low chlorinated congeners generally exhibited greater removal than the high ones.

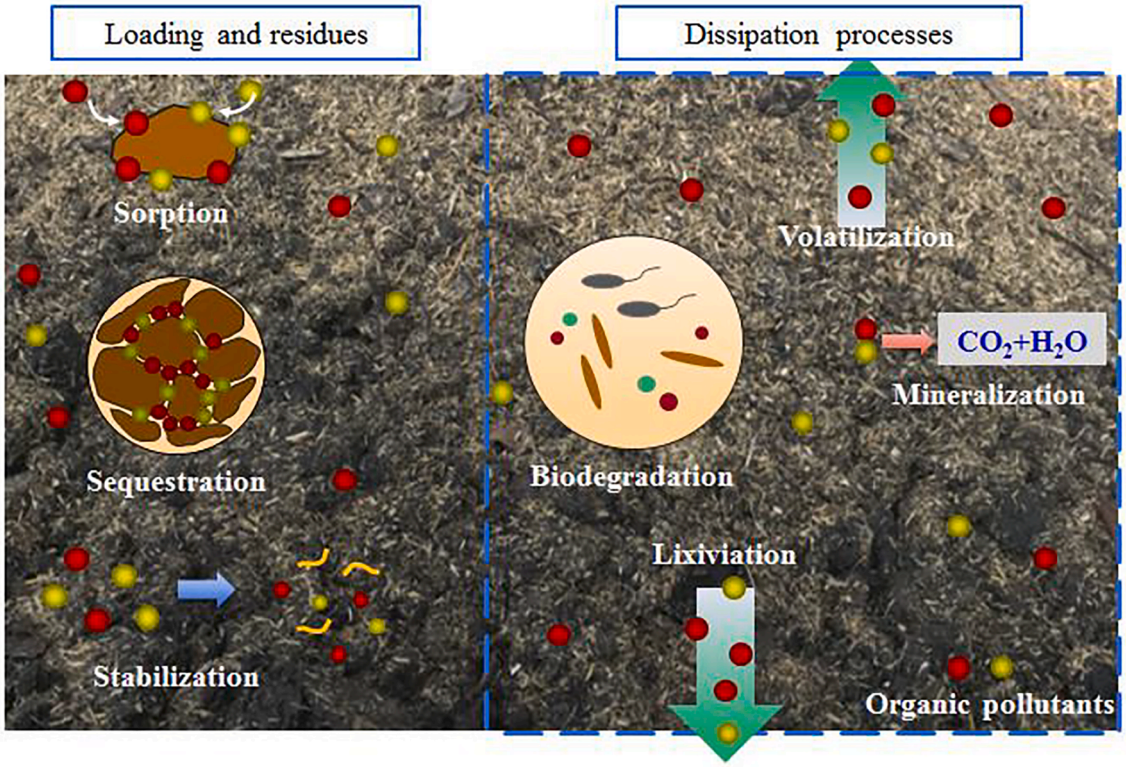

Fig. 2. Environmental behavior and fates of organic pollutants during sludge composting.

Biodegradation or mineralization is the primary pathway for removal or dissipation of organic pollutants during sludge composting (Amir et al., 2005b; Iranzo et al., 2018). Organic pollutants could be degraded/transformed into less complex intermediates, and then mineralized into inorganic minerals including H_2O, CO_2 (aerobic) or CH_4 (anaerobic) (Haritash and Kaushik, 2009). Lashermes et al. (2012) reported that mineralization (51% of initial LAS) contributed largely to the dissipation of ^{14}C-labeled LAS, whereas mineralization was moderate for ^{14}C-nonylphenol (29%) and ^{14}C-glyphosate (24%), and negligible for ^{14}C-fluoranthene during sludge composting with green waste. Microbial degradation of organic contaminants by mesophilic and thermophilic populations played a vital role in their removal during sludge composting (Amir et al., 2005a; Gibson et al., 2007; Liao et al., 2018). Degradation of organic micro-pollutants (e.g., DEHP and tri-n-butyl phosphate) was about 3-fold faster at early times than at later times, and the fastest degradation generally occurred at 35–60 °C of sludge - composting (Sadef et al., 2014). The biodegradation of organic contaminants and its efficiency are attributable to microbial genera/strains capable of degrading organic contaminants and their activities as well as physico-chemical properties of organic contaminants. Suitable temperatures promoted microbial growth and increased activities, especially for decomposing microorganisms and pollutant-degrading strains or consortium (Sadef et al., 2014; Yu et al., 2018b).

The degradation or mineralization of organic pollutants was significantly influenced by their bioavailability during sewage sludge composting (Oleszczuk, 2009; Aemig et al., 2019; Lu et al., 2019). Sorption or sequestration of organic contaminants by organic matter fractions influenced the bioavailability of organic contaminants, and thus lowered their dissipation during sludge composting (Oleszczuk, 2009; Lu et al., 2019). The effects of the composting process on PAH bioavailability depended on sewage sludge type, the composting methods and stages (Oleszczuk, 2009; Aemig et al., 2019; Lu et al., 2019). With the temperature decrease during composting, PAHs were stabilized in less or not accessible organic matter fractions (Aemig et al., 2019), and sequestration processes predominated for PAHs (especially 6-ring PAHs) in the maturation phase (Oleszczuk, 2009). At the end of sludge composting, the non-extractable residual proportions (% of recovered ^{14}C) were 45 ± 2 for 4-n-nonylphenol, 24 ± 7 for fluoranthene, and 14 ± 3 for LAS (Lashermes et al., 2012). Either irreversible sorption or physical sequestration might contribute to PAH decline at the end of composting (Amir et al., 2005a). Inconsistently, Aemig et al. (2019) reported that the chemical accessibility of PAHs increased in the maturation phase, because the amount of accessible organic matter fractions increased while the less accessible fractions decreased. Allowing adequate time for all composting phases could ensure efficient removal for most micro-pollutants (Sadef et al., 2014). In addition, bioleaching conditioning elevated significantly PAH bioavailability in dewatered sludge, and enhanced PAH dissipation during dewatered sludge composting (Lu et al., 2019). Nevertheless, limited information is available on the sorption/sequestration and bioavailability of other classes of organic contaminants during sludge composting.

Although some studies reported the dissipation pathways for several classes of organic contaminants, few studies have been emphasized on the removal mechanisms (e.g., biotic/abiotic dissipation, and bound residues formation) (Ezzariai et al., 2018). These removal percentages of organic contaminants were generally based on the differences of measured concentrations in initial and final composting mixtures, but the mass reduction (or material balance) was not taken into consideration (Ezzariai et al., 2018). In addition, many studies investigated only the concentration changes of parent compounds, while few studies analyzed their intermediates in composting mixture (Amir et al., 2005b), making it difficult to propose the degradation/mineralization pathway of organic contaminants during sludge composting. Moreover, certain intermediates (e.g., nonylphenols) exhibit higher toxicity to plants (e.g., wheat seedlings) than their parent compounds (e.g., nonylphenol ethoxylates) (Zhang et al., 2016b). The potential presence of more hazardous transformation products might influence the land application of sludge composts. Therefore, more investigation is required on removal pathways and mechanisms of recalcitrant organic contaminants.

4. Microbial community driving degradation of organic pollutants during composting

With the rapid development on analysis technology of microbial community, significant advances were gained on the succession and diversity of microbial community as well as their functions for degradation of organic pollutants (Alves et al., 2019; Guo et al., 2020). Previously, due to the limit of analysis technique, some studies generally analyzed microbial populations and microbial biomass carbon/nitrogen in sludge composting. Subsequently, the microbial communities in mixtures of sludge composts were analyzed using phospholipid fatty acids (Alves et al., 2019) or PCR-denaturing gradient-gel-electrophoresis (Nakasaki et al., 2009). Some recent studies have used high-throughput sequencing such as pyrosequencing and Illumina MiSeq platform to reveal the changes in the microbial community during sludge composting (Yu et al., 2018b; Ma et al., 2019; Guo et al., 2020). Microbial community of sludge composts was influenced by different composting methods and parameters (C/N ratio, moisture), in different composting phases (Zheng et al., 2018), with different bulking agent (Alves et al., 2019) and inoculation of matured compost (Ma et al., 2019). Correspondingly, the composition and diversity of microbial community, especially some dominant and key degrading genera, influenced the degradation/mineralization and fates of organic pollutants during sludge composting (Fig. 3).

Recently, several studies have focused on the influences of organic pollutants on microbial community and enzyme activities during sludge composting (Yang et al., 2017; Guo et al., 2020; Chen et al., 2021). Distinct shifts in bacterial community structure and composition were found during sludge composting (Su et al., 2015; Yu et al., 2018b; Liu et al., 2019). For example, in the sludge composting spiked with benzo[a]pyrene, Firmicutes was the most abundant phylum (with the typical thermophilic bacteria *Bacilli* and *Clostridia* being the dominant classes) in the first 12 d, while Actinobacteria, Bacteroidetes and Proteobacteria were the dominant phyla from 12 to 48 d of composting (Liu et al., 2019). With the increasing benzo[a]pyrene concentrations, the richness of Firmicutes decreased distinctly, while those of Actinobacteria and Proteobacteria increased remarkably (Liu et al., 2019). The presence of antibiotics (20 mg/kg) declined the richness and cellulase activity in the composting (Chen et al., 2021). Unfortunately, the effect mechanism of organic contaminants on microbial community and their interaction during sludge composting remain unclear.

Composting processes could accelerate the biodegradation/mineralization of organic pollutants because of great microbial diversity and activity as well as abundant substrates. Some studies attempted to emphasize on the correlation between biodegradation of organic contaminants and microbial community (Yang et al., 2017; Liu et al., 2019). Some bacterial (e.g., Pseudomonas, *Bacillus,* Mycobacterium, and Rhodococcus) and fungal species (e.g., white rot fungus*,* Phanerochaete chrysosporium, and Aspergillus) were commonly investigated PAE- and PAH-degraders(Haritash and Kaushik, 2009; González-Abradelo et al., 2019). Some dominant genera such as Nonomuraea, Cohnella, and Steroidobacter capable of degrading PAHs were found in sludge composting spiked with benzo[a]pyrene, which might contribute to benzo[a]pyrene removal (Liu et al., 2019). Yang et al. (2017) found that *Geobacter* and *Mycobacterium* involved in degradation of tetrabromobisphenol-A might contribute to dissipation of tetrabromobisphenol-A during sludge composting. During composting of hydrocarbon residue, sludge might exhibit a bioaugmentation or biostimulant because of its rich diversity and microbial biomass, promoting the removal of total petroleum hydrocarbon in composts (Alves et al., 2019). It should be noted that most studies deduced the bacterial community/genera responsible for degradation of organic contaminants mainly based on the previously reported degraders of specific organic compounds, or based on the correlation between the relative abundance of microbial community and the removal percentages of organic pollutants during composting (Yang et al., 2017; Liu et al., 2019; Guo et al., 2020). It is lack of the validation through the isolating strains from sludge compost to degrade organic contaminants, or through analyzing the genes/enzymes associated with biodegradation of organic contaminants during sludge composting.

Additionally, the microbial community and their biodegradation for organic pollutants were influenced by temperature, oxygen, pH, and nutrient accessibility of compost mixtures (Haritash and Kaushik, 2009; Yu et al., 2018b; Ma et al., 2019). Hyperthermophilic phase in a full-scale sludge composting was dominated by *Thermaceae* (35.5–41.7%), while thermophilic phase was predominated by *Thermaceae* (28.0–53.3%) and *Thermoactinomycetaceae* (29.9–36.1%) (Yu et al., 2018b). The relative abundance of phylum Firmicutes gradually declined with the decreasing temperature during the sludge composting (Liu et al., 2019). Increased degradation of organic contaminants were generally found at elevated temperatures compared to ambient temperature (Moeller and Reeh, 2003; Sadef et al., 2014). For instance, significant degradation of 4-nonylphenols was observed at composting temperatures 35–55 °C, and optimal degradation of LAS in sewage sludge was found at 55 °C (Moeller and Reeh, 2003). The rapidest removal of DEHP and triclosan was at moderate temperatures (30–50 °C), implying that some microorganisms might be active in degradation (Sadef et al., 2014). In certain case, high degradation rate of DEHP in the compost was only observed in a relatively narrow temperature range (about 37 °C), implying only a few DEHP-degraders present (Sadef et al., 2014). Unfortunately, the presence of antibiotics (especially at high concentration) deferred the appearance of the maximum temperature and obstructed the thermophilic phase (Ezzariai et al., 2018), which might affect the removal of other organic contaminants.

5. Research limitations and perspectives

Significant advance was obtained on the removal and dissipation mechanism of organic pollutants. The composting methods and processes were optimized to promote the removal and dissipation of organic pollutants. Nevertheless, there are some following knowledge gaps on organic pollutants during sludge composting which requires further research.

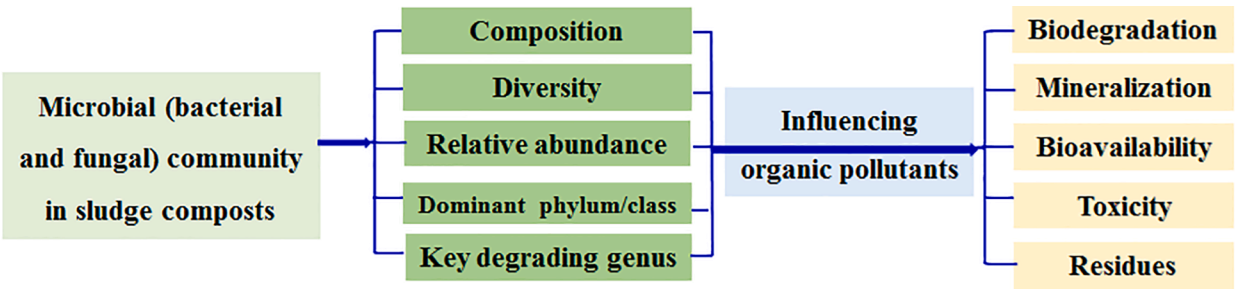

Fig. 3. Microbial community in sludge composts influencing environmental behavior and fates of organic pollutants.

(1) Sewage sludge contains many classes of organic pollutants with significant amounts, including some emerging organic pollutants such as organophosphate flame retardants, hormones and illicit drugs (Dubey et al., 2021). No literature to date addresses the occurrence and dissipation of these emerging pollutants during sludge composting.
(2) Many classes of organic pollutants coexist in sewage sludge, but very few comprehensive studies have reported the co-occurrence and simultaneous removal of different classes of organic pollutants during sludge composting (Cai et al., 2007c, 2012).
(3) Sludge composting can effectively remove some organic pollutants such as PAEs and PAHs. Nevertheless, some halogenated organic contaminants (e.g., PCDD/Fs, PCBs, and PFCs) are recalcitrant to be degraded during sludge composting, resulting in low removal or even elevated levels due to organic matter decomposition. Moreover, the disadvantage of long composting duration (e.g., >100 d) with low dissipation of organic pollutants limits the amount of sewage sludge to be composted. Great challenge exists for the balance among nutrient conservation, greenhouse gas reduction (Pan et al., 2018), and promoting dissipation of organic pollutants. Therefore, advanced pretreatment techniques and efficient composting process are needed for removing ARGs and recalcitrant organic pollutants and producing soil amendment.
(4) Many studies calculated the dissipation percentage of organic pollutants based on concentration differences at the beginning and end of sludge composting, without considering material balance, which might underestimate their removal efficiency.
(5) Many studies investigated only the parent compounds without giving information on their intermediates in composts. Analysis on intermediates of organic pollutants would be helpful for revealing their degradation/mineralization pathway during sludge composting.
(6) The current researches deduced the roles of microbial community in biodegradation mainly based on the previously reported degraders of specific organic pollutants, or the correlation between their dissipation percentages and the relative abundance of microbial community in composts. It needs to be proven through analysis of microbial genes/enzymes involving in pollutants degradation and/or isolating microbial consortium/strains from sludge composts to confirm the biodegradation of organic pollutants. Furthermore, revealing the interactions of microbial community and their influences on biodegradation of organic pollutants will be beneficial for elucidating their biodegradation mechanisms.

6. Conclusions

Sludge composts still contain various organic pollutants with the levels generally meeting the standard limits for land application. Sludge composting can effectively remove some organic pollutants such as PAEs and PAHs, while halogenated organic pollutants mainly remain in sludge composts with low dissipation rates or even at increased levels. The dissipation pathway including biodegradation or mineralization was elucidated for some organic pollutants. Moreover, sludge composting shifted distinctly microbial community in which some specific microbial classes/genera promote the degradation of organic pollutants. This systematical review would advance our understanding on dissipation of organic pollutants during sludge composting and promote the development of this field.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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